Dear editor and reviewers,

Thank you so much for your constructive comments in the discussion phase. Please check our responses to all your comments point by point (followed and marked in blue). Unless specified, all the line numbers in our responses refer to the line numbers of the TRACK CHANGES version of the manuscript.

Beside the responses to the two reviewers, we also made the following updates to the manuscript:

- We made grammatical edits throughout the manuscript.
- Due to a minor mistake in the simulations of our last version manuscript, we re-performed all the numerical simulations. The following changes / unchanged contents should be noted:

  1. Necessary updates were made to the text describing the new results.
  2. New results do not alter the main conclusions of this study. The new simulation results showed changes in the absolute values of N fluxes and loads. However, the variation patterns across scenarios (e.g., differences among WY, NY, DY, EDY) persisted. The effects of varying monthly average storm duration distributions and inter-storm period distributions on SON, SIN loads, leaching flux, and in-stream nitrate concentrations were also consistent. Therefore, the main conclusions of this study remain unchanged.
  3. New figures (Figure 3 - Figure 13) were re-regenerated based on the new simulation.
  4. Parameter $P_{drizzle}$ (probability of drizzle events) was additionally discussed as its effect on N dynamics were newly identified (lines 724-742, 883-894).

- We revised the document format according to the requirements.
- We added an authors' affiliation (Lines 8-9).

Responses:

RC1:

This manuscript presents a numerical modeling study exploring how inter- and intra-annual precipitation variability affects nitrogen (N) loads and fluxes in a catchment. The topic is timely and important for water quality and environmental management. The paper is generally well written, though some grammatical edits are needed. The methods and results are mostly clear and logically presented. The conclusions are relevant and likely to be of interest to scientists and resource managers focused on mitigating nutrient pollution.

Primary suggestions are to:

(1) Clarify and narrow the research focus: While the broad importance of precipitation variability is well established in the introduction, the final introductory paragraph should better define the unique contribution this work makes to the literature. Consider making the statement of objectives more specific and clarifying how the objectives fill gaps left by other recent studies.

Response #1:

Thanks for point that out! We have rephrased the last paragraph of the introduction part to define the unique contribution this work as "T*o fill the gap, the present study explored the impact of rainfall variability on N dynamics and its potential influence on water quality across inter-annual and intra-annual timescales. To characterize rainfall variability, a stochastic rainfall generator [Robinson and Sivapalan, 1997] was employed to generate rainfall time series with different climatic characteristics. The research was conducted in a small agricultural catchment located in Central Germany, where a hydrological model was previously established utilizing the fully coupled surface-subsurface numerical simulator HydroGeoSphere [Yang et al., 2018]. The framework of N dynamics was modified from the ELEMeNT approach [Exploration of Long-tErM Nutrient Trajectories; Van Meter et al., 2017]. The research is divided into two main components. First, three representative years (with high, normal, and low annual precipitation amounts, respectively) were chosen from the past two decades in Central Germany as the target scenarios. A fourth scenario with low annual precipitation amounts coupled with reduced plant uptake represents a case where vegetation is partially destroyed by extreme droughts. The statistical analyses of N loads and fluxes and a comparison across different scenarios were* *conducted to reveal the influence of inter-annual rainfall variability. Second, rainfall time series generated using the stochastic rainfall generator by separately altering specific parameters were used to substitute the rainfall data in the simulation period to drive the flow and nitrogen transport models. The responses of the N loads and fluxes to the parameters (e.g., the amplitudes of the seasonal variations in the storm duration and inter-storm period) were thoroughly analyzed to clarify the effect of intra-annual rainfall variability. The study will provide theoretical support for formulating fertilization strategies and protecting aquatic ecosystems in the context of climate change*" (Lines 133-163).

Reference:

Robinson, J., & Sivapalan, M.: Temporal scales and hydrological regimes: Implications for flood frequency scaling, Water Resour. Res., 33(12): 2981-2999, https://doi.org/10.1029/97WR01964, 1997.

Yang J., Heidbüchel, I., Musolff, A., Reinstorf, F., and & Fleckenstein. J. H.: Exploring the dynamics of transit times and subsurface mixing in a small agricultural catchment, Water Resour. Res., 54, https://doi.org/ 10.1002/2017WR021896, 2018.

Van Meter, K. J., Basu, N. B., Van Cappellen, P.: Two centuries of nitrogen dynamics: Legacy sources and sinks in the Mississippi and susquehanna river basins, Global Biogeochem. Cycles, 31 (1), 2–23., https://doi.org/10.1002/2016GB005498, 2017.

(2) Differentiate from previous work: The study references and builds on previous work, particularly Wang et al. (2023). However, the manuscript does not always make it clear where prior work ends and the current study begins. The manuscript also refers the reader to the previous studies for some crucial details (e.g. calibration to stream nitrate concentrations), which are not easy to find in the previous studies. I recommend adding further descriptions of the related studies at this site and delineating which analyses, model developments, and findings are new in this study.

Response#2:

Thanks for the points!

Generally, this study employed the model developed in our previous work Wang et al.

[2023] for a same study site. The model setup, method for simulating the flow and N transport and calibration, part of parameters are all from Wang et al. [2023]. However, new contributions are attributed to exploring the effect of rainfall variability on N dynamics. To clarify the existing work and the new contributions, we rephased the method section such that the new contributions are highlighted as follow: "*Based on the hydrological and transport model, the effect of topographic slope on the export of nitrate [Yang et al., 2022] and the spatial-temporal variation of nitrogen retention [Wang et al., 2023] were explored. In the present study, the two models were employed to investigate the effect of rainfall variability on N dynamics, for which rainfall time series with climatic characteristics, substituting for the precipitation data during the simulation period, were generated by a stochastic rainfall generator [Robinson and Sivapalan, 1997; Wilusz et al., 2017]*" (Lines 235-241).

Additionally, more details regarding the model calibration were added in the results section as a short review: "*The calibrated N transport model showed good performance in fitting the in-stream nitrate concentration (Figure 4a), with a Nash-Sutcliffe efficiency (NSE) of 0.79. The modeled N surplus of 51.87 kg ha^{-1} yr^{-1} is comparable to the measured value of 48.8 kg ha^{-1} yr^{-1}. The calibrated best-fit values for the transport parameters are listed in Table S1 (see Supporting Information)*" (Lines 487-491).

(3) Clarify model-data connection: A clearer connection could be made between the numerical model and real-world observations. The use of simple abstractions in the nutrient transport component of the model is reasonable, but also requires careful consideration of (a) how well the simplified processes representations mimic actual processes, (b) the uncertainty of the parameter estimates, and (c) the accuracy of the model in terms of reproducing observations. Otherwise, there is a risk of circularity: the model is built around certain processes and parameterizations, and then used to test the importance of those same processes and parameters.

Response#3:

Thanks for the suggestions. We made the following changes to clarify model-data connection:

1. We added the observed in-stream nitrate concentrations along with the simulated ones (see the figure below) such that they can be clearly compared.

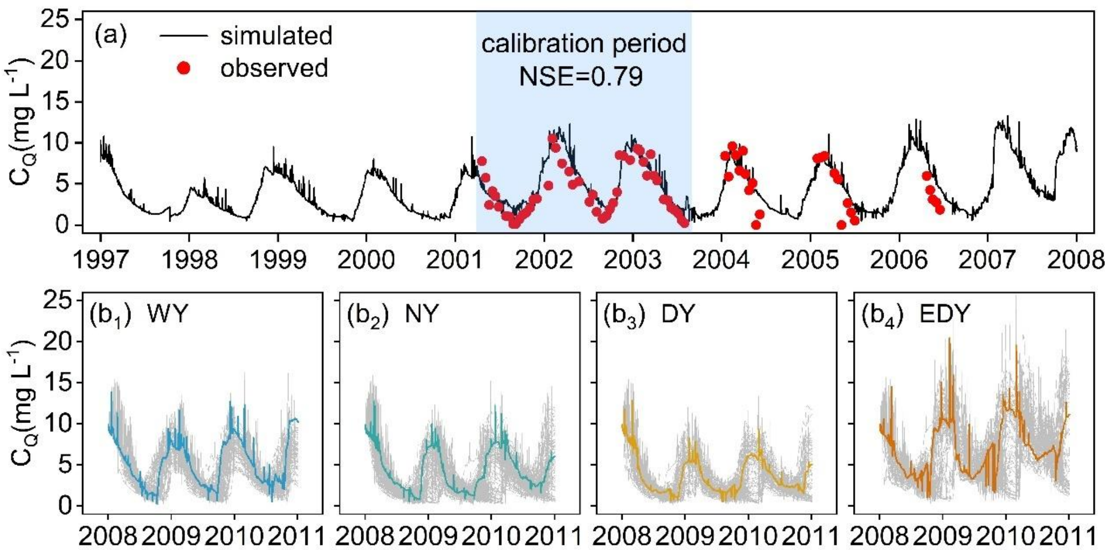

Figure 4. *The fluctuations of simulated in-stream nitrate concentration (C_Q) under (a) the historical rainfall (1997-2007) and the scenarios of (b_1) WY, (b_2) NY, (b_3) DY, and (b_4) EDY (2008-2010). The grey areas are formed by the realizations of each scenario.*

We added a description of the calibration results to show the accuracy of the model in terms of reproducing observations as "*The calibrated N transport model showed good performance in fitting the in-stream nitrate concentration (Figure 4a), with a Nash-Sutcliffe efficiency (NSE) of 0.79. The modeled N surplus of 51.87 kg ha^{-1} yr^{-1} is comparable to the measured value of 48.8 kg ha^{-1} yr^{-1}. The calibrated best-fit values for the transport parameters are listed in Table S1 (see Supporting Information). Figure 3 illustrates the 14-year N mass balance simulated by the calibrated N transport model in the entire catchment. In the soil source zone, the total N consisted of SON (552 kg ha^{-1}, including SON(a) of 90 kg ha^{-1} and SON(p) of 462 kg ha^{-1}) and SIN (48 kg ha^{-1}). The load of SON accounts for 92% of the total N, which corresponds to the research result that the organic N fraction is greater than 90% [Stevenson., 1995]. As for N transformation, the mineralization rate of 173 kg ha^{-1} yr^{-1} is within the range (14-187 kg ha^{-1} yr^{-1}) reported by Heumann et al. [2011] for study sites located in Germany. 71% of the SIN was absorbed by plants at a rate of 123 kg ha^{-1}*

yr^{-1}, which is very close to the value (around 120 kg ha^{-1} yr^{-1}) suggested in Nguyen et al. [2021] for the same area. The denitrification flux of 38 kg ha^{-1} yr^{-1} (4 kg ha^{-1} yr^{-1} in the soil source zone, 34 kg ha^{-1} yr^{-1} in the groundwater) is within the range (8-51 kg ha^{-1} yr^{-1}) investigated for 336 agricultural areas around the world by Hofstra and Bouwman [2005]. The SIN entered the groundwater zone at a rate of 44 kg ha^{-1} yr^{-1}, which is within the range (15-60 kg N ha^{-1} yr^{-1}) reported in Nguyen et al. [2021]. Approximately 23% of the leachate of SIN was exported to the stream, where the rest of the leachate (77%) was removed by denitrification. The export and denitrification fluxes are comparable to the values 39% and 61% reported in Nguyen et al. [2021]. Based on measured groundwater chemistry data from nearby catchments and multiple estimates of denitrification potential that have been proved and reported [Hannappel et al., 2018], we conclude that the simulated transformation and transport of N are acceptable.

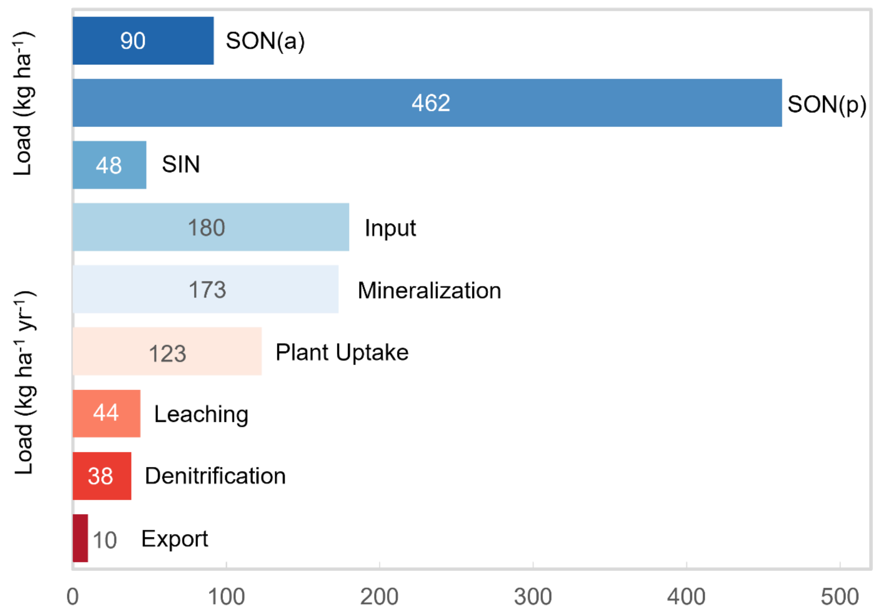

Figure 3. *The simulated N loads and fluxes in the entire catchment.*" (Lines 487-514).

2. We added short discussion about the uncertainty of the parameter in the section of Discussion as "*As a complex biogeochemical process, denitrification is governed by* various factors such as *temperature, soil moisture content, and SIN load,* rather than a first-order decay process. *Nonetheless, the current transport model does not account for spatial heterogeneity in denitrification.* These simplifications *may*

introduce uncertainties in the simulated results" (Lines 910-915).

Line-by-Line comments:

(4) Line 14: "performance" is wrong word. "effect"?

Response#4:

Thanks! We changed "performance" into "manifestation" (Line 17).

(5) 34: SON not defined at this point

Response#5:

Thanks, we rephrased as ".. soil organic nitrogen (SON)…" (Lines 41-42).

(6) 39 – "a small effect"

Response#6:

We corrected accordingly (Line 47).

(7) 58 – not clear what "their" refers to

Response#7:

We clarified it as "the effect of rainfall variability" (Line 79).

(8) 59 – grammar problem

Response#8:

We rephrased the sentence into "*anthropogenic amplification of rainfall variability has been identified [Zhang et al., 2024]*" (Lines 75-79).

(9) 72 –"a major"

Response#9:

We corrected accordingly (Line 95).

(10) 59-78 – the paragraph starts with climate variability and ends with nitrate. I recommend keeping to one topic per paragraph.

Response#10:

Thanks for the suggestion. We rephrased the content accordingly into three paragraphs (Lines 71-101).

(11) 99-100 sentence fragment

Response#11:

Thanks! The sentence was modified into "*Notably, the 2018 event triggered* *unprecedented tree mortality across multiple species in Central European forests, accompanied by unexpectedly persistent drought legacy effects [Schuldt et al., 2020]"* (Lines 112-114).

Reference:

Schuldt, B., Buras, A., Arend, M., Vitasse, Y., Beierkuhnlein, C., Damm, A., Gharun, M., Grams, T. E. E., Hauck, M., Hajek, P., Hartmann, H., Hiltbrunner, E., Hoch, G., Holloway-Phillips, M., Körner, C., Larysch, E., Lübbe, T., Nelson, D. B., Rammig, A., Rigling, A., Rose, L., Ruehr, N. K., Schumann, K., Weiser, F., Werner, C., Wohlgemuth, T., Zang, C. S., & Kahmen, A.: A first assessment of the impact of the extreme 2018 summer drought on Central European forests, Basic Appl. Ecol., 45, 86–103, https://doi.org/10.1016/j.baae.2020.04.003, 2020.

(12) 105 – "it is"

Response#12:

We corrected accordingly (Line 131).

(13) 107 – The objectives are somewhat broad. It has been established that precipitation variability can affect N dynamics, and it would help to be more specific in this paragraph about the aspects of variability being tested and what if anything has been done to address them previously. In other words, how the specific objectives of this study relate to gaps in knowledge left by previous studies?

Response#13:

Thanks for the suggestion. The paragraph was rephrased as "*To fill the gap, the present study explored the impact of rainfall variability on N dynamics and its potential influence on water quality across inter-annual and intra-annual timescales. To characterize rainfall variability, a stochastic rainfall generator [Robinson and Sivapalan, 1997] was employed to generate rainfall time series with different climatic characteristics. The research was conducted in a small agricultural catchment located in Central Germany, where a hydrological model was previously established utilizing the fully coupled surface-subsurface numerical simulator HydroGeoSphere [Yang et al., 2018]. The framework of N dynamics was modified from the ELEMeNT* *approach [Exploration of Long-tErM Nutrient Trajectories; Van Meter et al., 2017]. The research is divided into two main components. First, three representative years (with high, normal, and low annual precipitation amounts, respectively) were chosen from the past two decades in Central Germany as the target scenarios. A fourth scenario with low annual precipitation amounts coupled with reduced plant uptake represents a case where vegetation is partially destroyed by extreme droughts. The statistical analyses of N loads and fluxes and a comparison across different scenarios were conducted to reveal the influence of inter-annual rainfall variability. Second, rainfall time series generated using the stochastic rainfall generator by separately altering specific parameters were used to substitute the rainfall data in the simulation period to drive the flow and nitrogen transport models. The responses of the N loads and fluxes to the parameters (e.g., the amplitudes of the seasonal variations in the storm duration and inter-storm period) were thoroughly analyzed to clarify the effect of intra-annual rainfall variability. The study will provide theoretical support for formulating fertilization strategies and protecting aquatic ecosystems in the context of climate change*" (Lines 133-163).

(14) 163: cross section is not discernable and it is not clear what is the source of the saturation values

Response#14:

Thanks for pointing that out. We modified Figure 1 and its caption accordingly [Lines 205-212]:

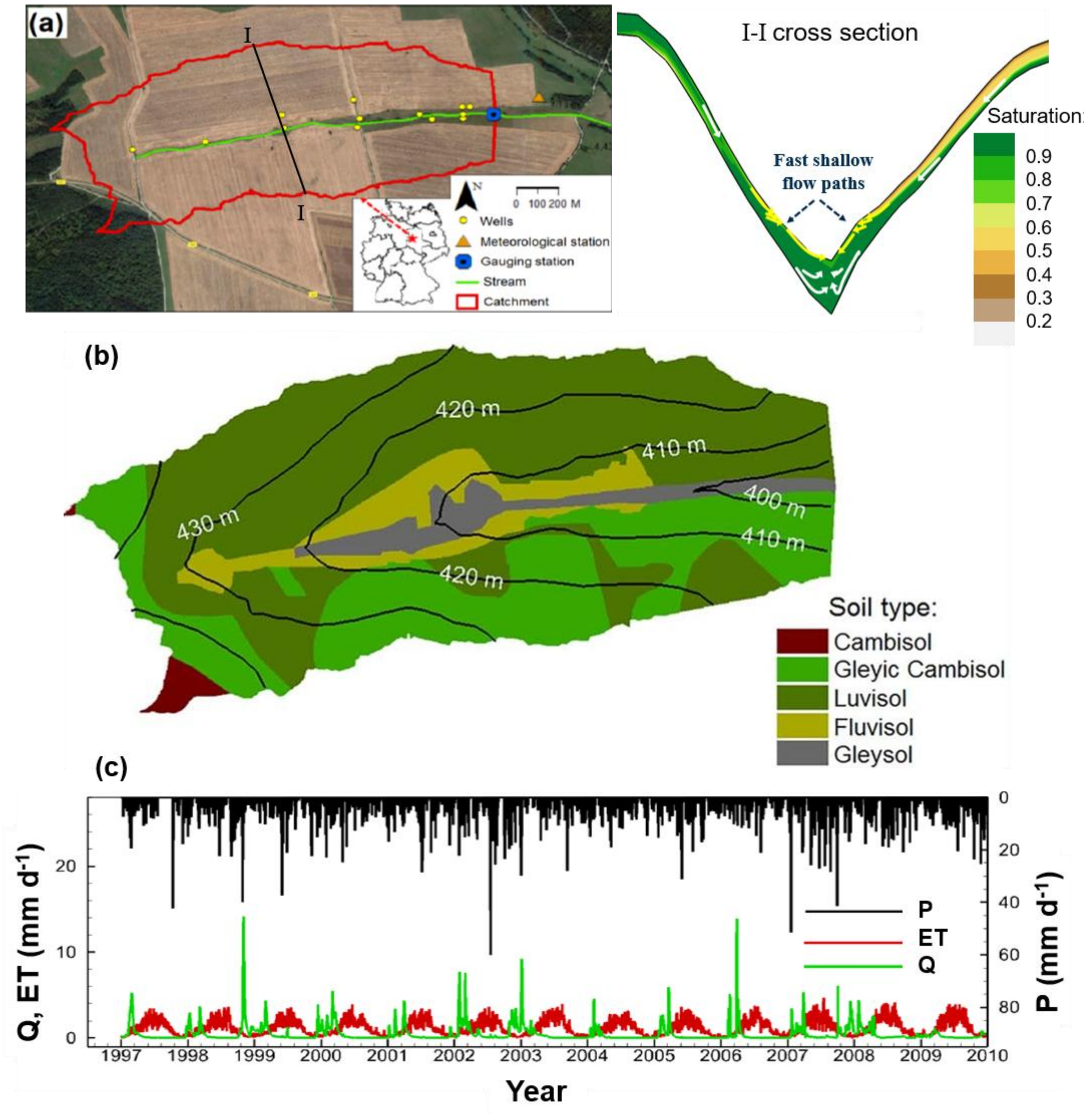

Figure 1. (a) The catchment 'Schäfertal' indicated by the red line (background image from © Google Maps), with a cross-sectional view for the flow and saturation [Yang et al., 2018]. (b) The distribution of soil type in the catchment. (c) The measured daily precipitation (P), discharge (Q), and the simulated actual evapotranspiration (ET) [Yang et al., 2018].

(15) 164: this figure is mostly recycled from Wang et al., 2023 but no citation is given.

Response#15:

We added the reference "*Yang et al. [2018]*" accordingly (Line 210).

(16) 169: Please clarify if/how these data are used in the current study.

Response#16:

Thanks for the suggestion.

We clarified that by adding "*The C_Q and N surplus are used to calibrate the N* *transport model [Wang et al., 2023]*" (Lines 225-226).

(17) 192 – delete "in details"

Response#17:

We corrected accordingly (Line 242).

(18) 231 – what does this mean that the calibrated model was "verified" over the entire simulation period?

Response#18:

Thanks! We rephrased the sentence into "*Subsequently, the calibrated model was verified by reproducing time-variable groundwater levels for the wells over the entire simulation period [Yang et al., 2018]*" (Lines 283-284).

(19) 240 – meaning unclear "delineated corresponding to the reality"

Response#19:

Thanks! We have removed the unnecessary sentences (Lines 290-296).

(20) 245-6: "in route" grammar

Response#20:

Thanks for pointing that out. The sentence was modified to "*Throughout the nitrogen cycle, various forms of nitrogen undergo complex biogeochemical processes*" (Lines 299-300).

(21) 258-260: meaning unclear.

Response#21:

We rephrased the sentence into "*The framework is able to capture the main processes of nitrogen transformation and transport in soil and groundwater [Yang et al., 2018]*" (Lines 312-316).

(22) 320: "validation" might not be the right word (not the same as calibration).

Response#22:

Thanks for pointing that out. This related sentence was deleted. The brief calibration results were added in the beginning of result section as "*The calibrated N transport model showed good performance in fitting the in-stream nitrate concentration (Figure 4a), with a Nash-Sutcliffe efficiency (NSE) of 0.79. The modeled N surplus of 51.87 kg*

ha^{-1} yr^{-1} is comparable to the measured value of 48.8 kg ha^{-1} yr^{-1}. The calibrated best-fit values for the transport parameters are listed in Table S1 (see Supporting Information)" (Lines 487-491).

(23) 323: Wang et al 2023 refers readers to Yang 2018 for more details and is not an easy source of information about the estimation of the N cycling parameters, uncertainty of those parameters or the quality of fit to the data. These are crucial aspects of the calibrated model and should be presented clearly and succinctly for the readers.

Response#23:

Thanks for the suggestion. The calibration result was added in the beginning of result section as "*Figure 3 illustrates the 14-year N mass balance simulated by the calibrated N transport model in the entire catchment. In the soil source zone, the total N consisted of SON (552 kg ha^{-1}, including SON(a) of 90 kg ha^{-1} and SON(p) of 462 kg ha^{-1}) and SIN (48 kg ha^{-1}). The load of SON accounts for 92% of the total N, which corresponds to the research result that the organic N fraction is greater than 90% [Stevenson., 1995]. As for N transformation, the mineralization rate of 173 kg ha^{-1} yr^{-1} is within the range (14-187 kg ha^{-1} yr^{-1}) reported by Heumann et al. [2011] for study sites located in Germany. 71% of the SIN was absorbed by plants at a rate of 123 kg ha^{-1} yr^{-1}, which is very close to the value (around 120 kg ha^{-1} yr^{-1}) suggested in Nguyen et al. [2021] for the same area. The denitrification flux of 38 kg ha^{-1} yr^{-1} (4 kg ha^{-1} yr^{-1} in the soil source zone, 34 kg ha^{-1} yr^{-1} in the groundwater) is within the range (8-51 kg ha^{-1} yr^{-1}) investigated for 336 agricultural areas around the world by Hofstra and Bouwman [2005]. The SIN entered the groundwater zone at a rate of 44 kg ha^{-1} yr^{-1}, which is within the range (15-60 kg N ha^{-1} yr^{-1}) reported in Nguyen et al. [2021]. Approximately 23% of the leachate of SIN was exported to the stream, where the rest of the leachate (77%) was removed by denitrification. The export and denitrification fluxes are comparable to the values 39% and 61% reported in Nguyen et al. [2021]. Based on measured groundwater chemistry data from nearby catchments and multiple estimates of denitrification potential that have been proved and reported [Hannappel* *et al., 2018], we conclude that the simulated transformation and transport of N are acceptable.*

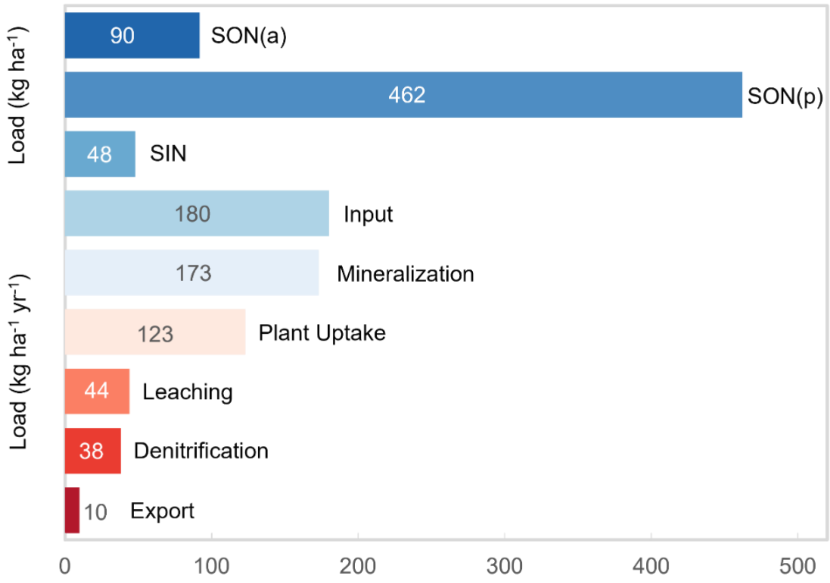

Figure 3. *The simulated N loads and fluxes in the entire catchment.*" (Lines 492-514).

(24) 325: "impermeable for nitrate" (and water?)

Response#24:

We rephrased the sentence as "*The bedrock is treated as impermeable for water and nitrate*" (Lines 384-385).

(25) 333-375: probably don't need this much detail about the rainfall generator

Response#25:

Thanks for the suggestion. The section 3.3 has been simplified (Lines 393-442).

(26) 336: "a stochastic model"

Response#26:

We corrected accordingly (Line 395).

(27) Table 1 – This table has too many numbers and variables for readers to easily absorb. Consider placing with a schematic, examples, or another simpler figure or table.

Response#27:

Thanks for the suggestion. We updated Table 1 accordingly.

(28) 399-401 – grammar problem, meaning is lost

Response#28:

Thanks for pointing that out. The sentence has been modified to "*In order to explore the effect of intra-annual rainfall variability on N dynamics, the linear regression analyses between the parameters of the stochastic rainfall generator and N loads, fluxes, as well as* C_Q *were conducted, in which the parameters of NY serve as a reference*" (Lines 467-470).

(29) 458 – Why is soil denitrification lumped with GW denitrification? Are they expected to be similar?

Response#29:

Thanks for point that out. Soil denitrification and GW denitrification are identical in mechanism, and we expected to see how much nitrogen in total is consumed by denitrification under different rainfall pattern. Therefore, they are added together in the study.

(30) 503 – These figures are confusing because the response variables (SON, SIN, LEA, Cq) are not on the z-axes.

Response#30:

Thanks for the suggestion! Figure 9 and Figure 10 were presented in the same way. In the stochastic rainfall generator, the distributions of the average storm duration and the average inter-storm period over the course of a year are depicted by Sine functions (Equation S3 & S4, Supporting Information), whose characteristics are determined by the amplitudes of the seasonal variations in the average storm duration and the average inter-storm period (α_γ and α_δ) reflecting climate change, when other parameters remain constant. We will use Figure 9 as an example. As for a certain α_γ, there is a specific distribution of the average storm duration over the course of a year, which is presented in the x-z plane. The color of the panels represents the value of α_γ. With α_γ decreasing, the distribution of the average storm duration transitions and the year shifts from wet to dry. When the year becomes drier, the annual SON load increases, while the SIN load, leaching flux, and in-stream nitrate concentration decrease on the y-axis. Therefore, the transformation and transport of N are subject to retardation in a dry year when storms with shorter duration occur mid-year.

(31) 564: 5.1 section title: consider being more specific about what increased rainfall does to the N dynamics

Response#31:

Thanks for the suggestion. We changed section tile of 5.1 into "*N transformation and transport upgrade in wet years*" (Lines 745-746).

(32) 608: It seems notable that the high flows during the 2018 to 2019 drought are as high as the high flows from 2014-2018, and the main difference during 2018-2019 seems to be in the low flow periods.

Response#32:

Thanks for pointing that out. Yes, as you observed, droughts mainly occurred in the middle of the year, resulting in extreme low flow (Figure 14). During the low-flow periods, nitrogen absorbed by vegetation in the growth stage is significantly reduced and nitrogen (SON & SIN) starts to accumulate in the soil. The accumulated nitrogen was subsequently flushed out during the rewetting period [Winter et al., 2023].

Reference:

Winter, C., Nguyen, T., Musolff, A., Lutz, S., Rode, M., Kumar, R., Fleckenstein, J.: Droughts can reduce the nitrogen retention capacity of catchments, Hydrol. Earth Syst. Sci., 27(1):303-318, https://doi.org/10.5194/hess-27-303-2023, 2023.

(33) 670: 5.6. Consider discussing: Data limitations, uncertainty of parameters, model process representations

Response#33:

Thanks for the suggestions.

We added the discussion to address the model representation and parameter uncertainty and limitation in data as "*The transport model preserves the main pathway for N-NO_3^- leachate by simplifying complexities of different N pools and transformations via mineralization, leaching, and denitrification within the soil zone. However, the external N input entering directly into inorganic N, the transformation from protected organic N to active organic N, and the loss of organic N via dissolution have not been included in the model, which may lead to miscalculation of* *nitrogen load. As a complex biogeochemical process, denitrification is governed by various factors such as temperature, soil moisture content, and SIN load, rather than a first-order decay process. Nonetheless, the current transport model does not account for spatial heterogeneity in denitrification. These simplifications may introduce uncertainties in the simulated results. In addition, due to the lack of groundwater nitrate concentration data, the nitrate transport model was calibrated using only in-stream nitrate concentration data [Wang et al., 2023]. More nitrate concentration data in groundwater and river will contribute to establishing a more realistic nitrate transport model*" (Lines 905-919).

(34) Table S1 – van Meter reference is missing date; bibliography is not included in this document

Response#34:

Thanks for pointing that out. We have corrected the table S1:

Table S1. The parameters for the N source zone. The parameters were adjustable and calibrated (referring to Yang et al. [2022]).

Parameter	Process	Adjustable range	Reference	Value of best fit
k_a	Mineralization	0~0.7	Van Meter et al. [2017]	0.0111 day^{-1}
k_p	Mineralization	0~0.7	Van Meter et al. [2017]	0.0008 day^{-1}
$p1$	Plant uptake	60~160	Van Meter et al. [2017]	160 kg·ha^{-1}
$p2$	Plant uptake	1~10		10 kg·ha^{-1}
$p3$	Plant uptake	1~60		34 day
λ_s	Denitrification in soil	0~0.7	Nguyen et al. [2021]	0.0007 day^{-1}
k_l	Leaching	1~1000		18.8888 day^{-1}
q_{ref}	Leaching	$1e^{-4}$~1		0.01 m·day^{-1}
λ	Denitrification in water	$1e^{5}$~$1e^{-1}$	Nguyen et al. [2021]	0.0088 day^{-1}

RC2:

The authors present an original piece of research focusing on the catchment response of N fluxes to rainfall interannuel and intra-annual variability using synthetic experiments based on the Hydrogeosphere model. I found the study clean and rigorously described, calibration method is sensible. Figures provide useful and clear illustrations of the results. The manuscript is well written. I think the discussion could be expanded and therefore I recommand a minor revision of the manuscript.

Especially I would have **2 comments on the discussion**:

1) several parts of the discussion present some reactions as simulation results when they are a direct consequence of the modelling equations. Ex.: Lines 566 to 568, the impact of temperature and wetness on mineralization is constructed mathematically in equations 1 and 2, isn't it? Same comment regarding plant uptake and denitrification. According to my opinion, the interest of the model is rather to calculate which of these mechanisms is going to dominate the response, and also it helps to consider different time scales of response, which is particularly relevant for droughts (cf. lines 97-99)

Response #1:

Thank you for the comment. We truly agree that the reaction is a direct consequence of the mechanisms that are built in the equations already. That mean, some trends / patterns can be simply known even without actually performing the simulations. This study is aimed to investigate how the rainfall variability influences the N transport and which process is the key control. E.g., we found that the plant-uptake is the main factor caused the high in-stream concentration in extreme dry years.

In response to your comment, we added the sentence in the result section "*Based on these results, it can be preliminarily concluded that annual precipitation and vegetation state critically influence N transformation and transport*" (Lines 590-591).

In addition, we have revised the discussion to clarify the dominant mechanisms governing in-stream nitrate concentration responses as "*The comparison of* C_Q*, N loads, and fluxes across four scenarios (WY, NY, DY, and EDY) reveals the effect of inter-annual rainfall variability on N transformation and transport, as well as water quality. Mineralization, the crucial process of the transformation from SON to SIN, exhibits strong soil moisture dependence. Consequently, the highest average mineralization rates in the WY scenario promoted N transformation through plant uptake, denitrification, and leaching. In contrast, low mineralization in the DY scenario led to SON accumulation, thereby restraining overall N dynamics*" (Lines 747-754).

2) I have a general comment on the modelling choice that is N mechanisms are much more simplified than the water processes. While I am very aware of the computationnal challenges associated with such virtual experiments, I find intellectually disturbing to have a fully mechanistic approach to represent water combined with a representation of nitrogen very simplified in comparison. What do you think?

Response #2.1:

Thank you for this comment! We totally agree with your point that the N processes represented in the model is very simple compared with the groundwater flow. The complexities N fluxes in source zone were simplified by defining a framework describing the main pathway for N-NO_3 leachate with temporally constant external N input. On the one hand, this simplification neglected other processes such as the time-variant external N input, direct input of external N to inorganic N pool, the transformation from protected organic N to active organic N, and the loss of organic N via dissolution. On the other hand, this simplification allows us to focus on the main source of N-NO_3^-, rather than keeping track of the full nitrogen fluxes in the source zone of the catchment while maintaining the overall nitrogen balance using surplus as a constraint. In this sense, we think it is an effective tool to answer certain questions, for example in our study, which process controls the high nitrate concentrations in river while dry climate.

To response to this point, we discussed the limitation of the simplified N framework additionally in section 5.7 as follow: "*The transport model preserves the main pathway for N-NO$_3^-$ leachate by simplifying complexities of different N pools and transformations via mineralization, leaching, and denitrification within the soil zone. However, the external N input entering directly into inorganic N, the transformation from protected organic N to active organic N, and the loss of organic N via dissolution have not been included in the model, which may lead to miscalculation of nitrogen load*" [Lines 905-910].

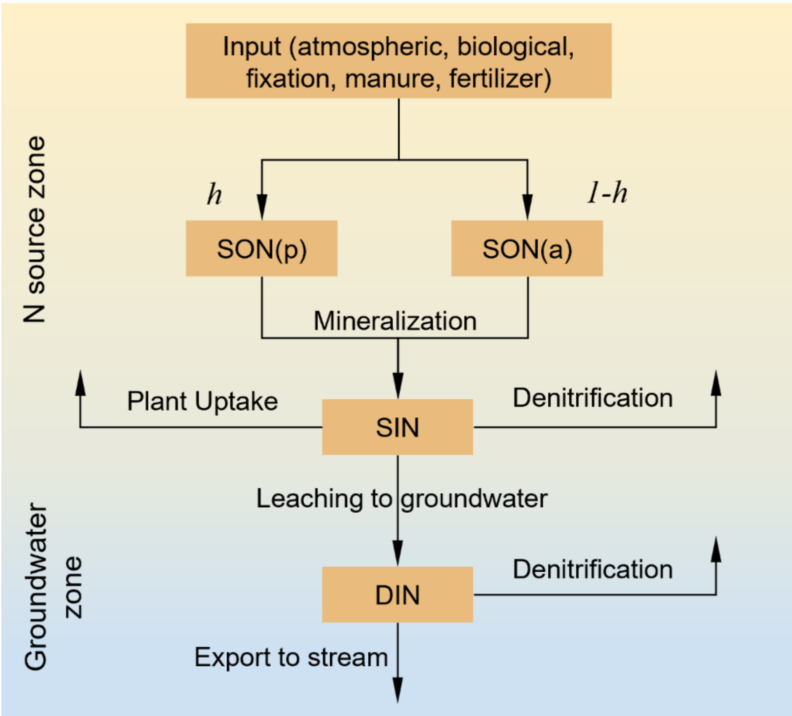

Figure 2. *The framework simulating the transformation and transport of nitrogen in soil N source zone and groundwater zone, modified based on [Yang et al., 2021].*

There is no representation of plant-soil processes, and considering the results of the EDY scenario in terms of N dynamics, the response of plants growth or death to water stress seems to be a key mechanism.

Response #2.2:

Thanks for pointing that out! Yes, the plant-soil process is not represented in the model. This is, in the extreme dry scenario, the plants death was not actually simulated under low soil content, but manually being assigned a lower plant uptake potential. The plant growth stages were assumed to be constant (for example, the same N uptake potential in different years). This is also due to lack of representation for the plant-soil processes in the simplified N framework. N uptake process is only mathematically described using the empirical formula (Equation 6). However, such simplification was proved to be effective in term of reproducing the reasonable N loads, fluxes and in-stream nitrate concentrations. It can still be used to identified the key effect of plant-die-off (i.e. reduced N uptake potential) on stream water quality during extreme dry climate, comparing with other process such as mineralization and leaching.

To response to this point, we discussed the limitation of neglecting the plant-soil processes in section 5.7 as follow: "*The plant-soil process was not represented in the model. In the extreme dry scenario, plant death was not actually simulated under high temperature and low soil moisture content, but was manually assigned a lower plant uptake potential. The plant growth stages were assumed to be constant. This is because the N uptake process was only mathematically described using the empirical formula (Equation 6) in the simplified N framework, rather than using a full plant-soil process. However, such simplification was proved to be effective in terms of reproducing reasonable N loads, fluxes and in-stream nitrate concentrations. Therefore, we think it is acceptable to identify the key effect of plant die-off (i.e. reduced N uptake potential) on stream water quality during extreme dry scenarios*" (Lines 924-934).

Also, about the fact that all external nitrogen inputs are introduced in the SON pool (lines 250, 252), I was wondering to what extend it refers to a reality? Are fertilizers mainly applied as urea?

Response #2.3:

Thanks for pointing that out!

The external N input represents atmospheric deposition, biological fixation, animal manure from the pasture area, and N fertilizer from the farmland. It does include organic and inorganic inputs. In our framework based on ELEMeNT model [Van Meter et al., 2017], it is assumed that all external nitrogen inputs are introduced in the organic pool. This assumption is made based on the fact that most of the nitrate (N-NO_3^-) fluxes from source zone has undergone biogeochemical transformation in organic N pool [Haag and Kaupenjohann, 2001]. The described framework simplifies complexities of different N pools and transformations via mineralization, dissolution, and denitrification within the soil zone, while preserving the main pathway for N-NO_3 leachate. In this sense, assumption that the external N input contributes only to the SON is acceptable in the study.

To response to this point, we discussed the limitation of the simplified N framework additionally in section 5.7 as "*The transport model preserves the main pathway for N-NO_3^- leachate by simplifying complexities of different N pools and transformations via mineralization, leaching, and denitrification within the soil zone. However, the external N input entering directly into inorganic N, the transformation from protected organic N to active organic N, and the loss of organic N via dissolution have not been included in the model, which may lead to miscalculation of nitrogen load. As a complex biogeochemical process, denitrification is governed by various factors such as temperature, soil moisture content, and SIN load, rather than a first-order decay process. Nonetheless, the current transport model does not account for spatial heterogeneity in denitrification. These simplifications may introduce uncertainties in the simulated results. In addition, due to the lack of groundwater nitrate concentration data, the nitrate transport model was calibrated using only in-stream nitrate concentration data [Wang et al., 2023]. More nitrate concentration data in groundwater and river will contribute to establishing a more realistic nitrate transport model*" (Lines 905-919).

Also, I did not understand why is this framework acceptable and according to which criteria or arguments (line 258-260)?

Response #2.4:

Thanks for the comment!

Although the mechanisms of N dynamics in this framework are quite simplified, its acceptability is supported by the following points:

- It is effective in term of reproducing the reasonable in-stream nitrate concentrations with higher NSE in the entire catchment. We added the calibration results in the beginning of the section 4 as follows: "*The calirbated N transport model showed good performance in fitting the in-stream nitrate concentration (Figure 4a), with a Nash-Sutcliffe efficiency (NSE) of 0.79. The modeled N surplus of 51.87 kg ha^{-1} yr^{-1} is comparable to the measured value of 48.8 kg ha^{-1} yr^{-1}. The calibrated best-fit values for the transport parameters are listed in Table S1 (see Supporting Information)*" (Lines 487-491).
- The described framework simplifies complexities of different N pools and transformations via mineralization, dissolution, and denitrification within the soil zone, while preserving the main pathway for N-NO_3^- leachate. In this sense, the framework can still be used to explore the influence of rainfall on water quality via the key processes (mineralization, plant-uptake and leaching).

To response this point, we briefly compared the simulated N loads and fluxes with observations or data from other literatures in the beginning of the section 4 as follows: "*Figure 3 illustrates the 14-year N mass balance simulated by the calibrated N transport model in the entire catchment. In the soil source zone, the total N consisted of SON (552 kg ha^{-1}, including SON(a) of 90 kg ha^{-1} and SON(p) of 462 kg ha^{-1}) and SIN (48 kg ha^{-1}). The load of SON accounts for 92% of the total N, which corresponds to the research result that the organic N fraction is greater than 90% [Stevenson., 1995]. As for N transformation, the mineralization rate of 173 kg ha^{-1} yr^{-1} is within the range (14-187 kg ha^{-1} yr^{-1}) reported by Heumann et al. [2011] for study sites located in Germany. 71% of the SIN was absorbed by plants at a rate of 123 kg ha^{-1} yr^{-1}, which is very close to the value (around 120 kg ha^{-1} yr^{-1}) suggested in Nguyen et al. [2021] for the same area. The denitrification flux of 38 kg ha^{-1} yr^{-1} (4 kg ha^{-1} yr^{-1} in the soil source zone, 34 kg ha^{-1} yr^{-1} in the groundwater) is within the range (8-51 kg ha^{-1} yr^{-1}) investigated for 336 agricultural areas around the world by Hofstra and*

Bouwman [2005]. The SIN entered the groundwater zone at a rate of 44 kg ha^{-1} yr^{-1}, which is within the range (15-60 kg N ha^{-1} yr^{-1}) reported in Nguyen et al. [2021]. Approximately 23% of the leachate of SIN was exported to the stream, where the rest of the leachate (77%) was removed by denitrification. The export and denitrification fluxes are comparable to the values 39% and 61% reported in Nguyen et al. [2021]. Based on measured groundwater chemistry data from nearby catchments and multiple estimates of denitrification potential that have been proved and reported [Hannappel et al., 2018], we conclude that the simulated transformation and transport of N are acceptable.

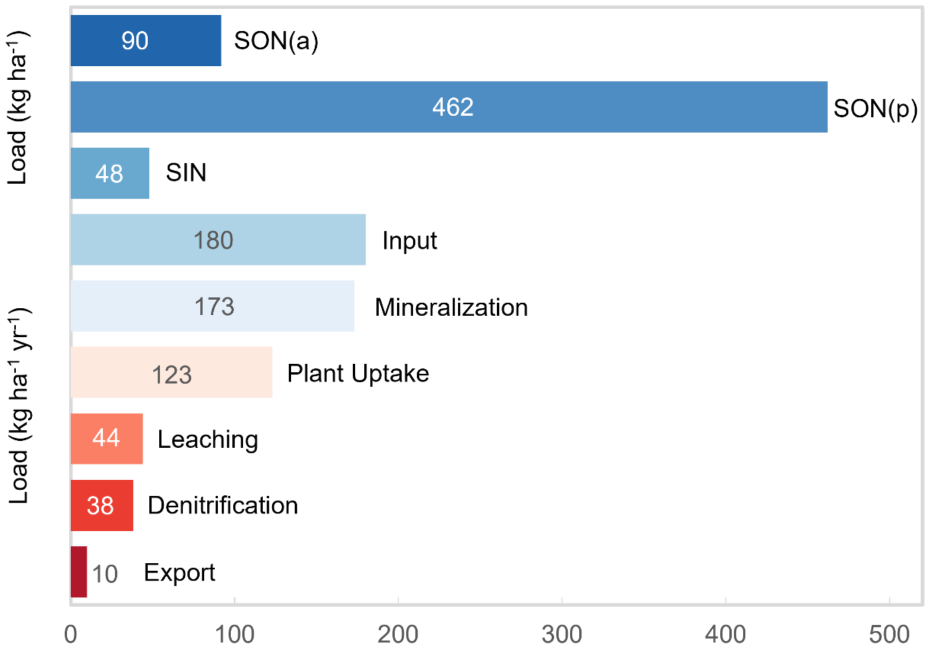

Figure 3. *The simulated 14-year N mass balance in the entire catchment.*" (Lines 492-514).

I have also a few minor comments below:

3) line 41: I suspect that fertilization strategies will evolve with the changes in timing of plant growth stages with warming

Response #3:

Thanks! This is a very good point. We agree that the fertilization strategies may evolve with warming. Due to the lack of spatiotemporal variation information of the external N input, its value was fixed at 180 kg ha^{-1} $year^{-1}$ according to Nguyen et al. [2021], where the N balance was simulated in the upper Selke catchment covering the Schäfertal catchment. In addition, in this study, the plant growth stages were assumed to be the same for different years (for example, the same sowing date in either wet year or dry year). This is due to lack of representation for the plant-soil processes in the model. Such assumption was proved to be effective in term of reproducing the reasonable N loads, fluxes and in-stream nitrate concentrations [Yang et al., 2021]. However, these assumptions may lead to uncertainties in the simulated N fluxes which is discussed in section 5.7 as following: "*The plant-soil process was not represented in the model. In the extreme dry scenario, plant death was not actually simulated under high temperature and low soil moisture content, but was manually assigned a lower plant uptake potential. The plant growth stages were assumed to be constant. This is because the N uptake process was only mathematically described using the empirical formula (Equation 6) in the simplified N framework, rather than using a full plant-soil process. However, such simplification was proved to be effective in terms of reproducing reasonable N loads, fluxes and in-stream nitrate concentrations. Therefore, we think it is acceptable to identify the key effect of plant die-off (i.e. reduced N uptake potential) on stream water quality during extreme dry scenarios*" (Lines 924-934).

4) introduction should maybe be restricted to context elements that are directly linked to the study, lines 45 to 61: it could go more directly to the point of rainfall variability skipping the details on extremes. Lines 79 to 82: I missed the link with the present study here.

Response #4:

Thank you for your suggestions. We have revised the first paragraph of the Introduction as "*The hydrological processes are susceptible to meteorological conditions on various spatial and temporal scales [Ionita et al., 2017; Laaha et al., 2017; Zhang et al., 2021]. In the past decades, extreme climate events intensified by human-induced climate change have frequently occurred globally [Pall et al., 2011;*

Min et al., 2011; Williams et al., 2015; Hari et al., 2020], most of which caused water scarcity and poor water quality at regional scales [Zwolsman and van Bokhoven, 2007; Delpla et al., 2009; Whitehead et al., 2009; Stahl et al., 2016; Ballard et al., 2019; Bauwe et al., 2020; Geris et al., 2022]. Heavy rainstorms and severe droughts being the predominant extreme climate events around the globe share the common characteristic of rainfall variability [Trenberth et al., 2011; Pendergrass et al., 2017; Hanel et al., 2018]. In the context of global warming scenarios, anthropogenic amplification of rainfall variability has been identified [Zhang et al., 2024]. Thus, the effect of rainfall variability on water resources has attracted much attention around the world" (Lines 59-80).

We also deleted the following sentences describing historical extreme events, which are not directly relevant to this study "*Extreme rainfall events in 2002, 2013, and 2021 caused significant threats to human safety and substantial damage to the environment, economy, and infrastructure [Ulbrich et al., 2003; Thieken et al., 2016; Voit & Heistermann, 2024]. Three notable summer droughts occurred in 2003, 2015, and 2018–2019 (consecutive), driven by precipitation deficits combined with temperature anomalies during the growing season [Fink et al., 2004; Schär et al., 2004; Ciais et al., 2005; Orth et al., 2016; Hanel et al., 2018; Hari et al., 2020; Camenisch et al., 2020]*" (Lines 104-111).

5) lines 145-146: at which frequency are concentration measured?

Response #5:

Thanks! We clarified this by adding "*..at 14-days to monthly intervals [Dupas et al., 2017], covering the period 2001–2010*" (Lines 186-187).

6) line 339-340 what is a "good performance under Climate change" for the stochastic generator?

Response #6:

Thank you for raising this point.

Wilusz et al. [2017] coupled the rainfall generator [Robinson & Sivapalan, 1997], a rainfall-runoff model [Kirchner, 2009], and the rank StorAge Selection (rSAS) transit time model [Harman, 2015] to decompose the relationship between rainfall variability and the time-varying fraction of young water (<90 days old) in streams (FYW). The rainfall generator was specifically employed to synthesize daily rainfall time series with statistical properties that are identical with or systematically different from historic rainfall. Crucially, the generator is capable of representing individual storm events, between-storm variability, within-storm variability, and seasonality [Robinson & Sivapalan, 1997]. Thus, based on these capabilities, the sentence was modified into "*It can output rainfall series representing different rainfall patterns under climate change*" (Lines 398-400).

7) Figure 4 a: it would be useful to have measured C_Q in the plot too. The legend refers to "acceptable simulations" but so far as I understand it is more the variability associated to the generator in each scenarios (n=100) isn't it?

Response #7:

Thank you for the suggestion!

We have added the measured in-stream nitrate concentration data to Figure 4a. Indeed, gray areas in Figure 4b_1~b_4 are formed by the 100 realizations. We changed the caption of Figure 4 to clarify this as "*The grey areas are formed by the realizations of each scenario*" (Lines 525-526).

8) lines 577-578: are preferential flowpaths represented in the model?

Response #8:

Thank you! Preferential flow paths on top of the aquifer (fast shallow flow paths) identified and discussed in our previous study [Yang et al., 2018], have been plotted in I-I cross section (Figure 1a). Such fast shallow flow paths established a hydraulic connection between channel and the slope top in the wet seasons. We added "*(I-I cross section, Figure 1a)*" (Lines 768-769) to refer to the flow paths.

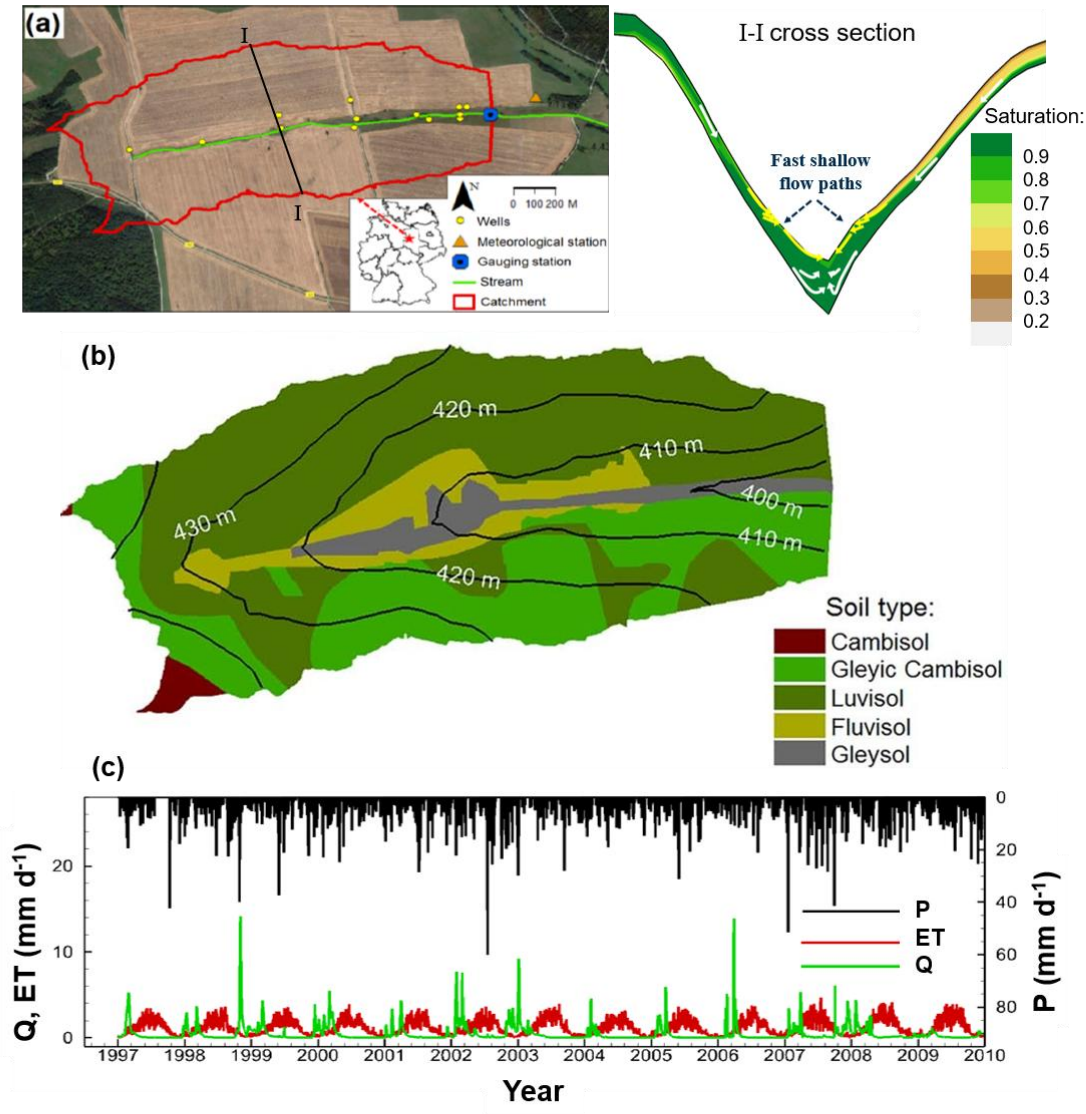

Figure 1. (a) The catchment 'Schäfertal' indicated by the red line (background image from © Google Maps), with a cross-sectional view for the flow and saturation [Yang et al., 2018]. (b) The distribution of soil type in the catchment. (c) The measured daily precipitation (P), discharge (Q), and the simulated actual evapotranspiration (ET) [Yang et al., 2018].

9) it would be useful to see the effect on water fluxes as well: especially recharge flux for groundwater and Actual ET in Figure 10 or Figure S2 (lines 653 to 656)

Response #9:

Thank you for your suggestion. We added the actual ET and the rechange fluxes (Rainfall-actual ET) in Figure S2 (see Supporting Information).